PATENT COOPERATION TREATY

From the INTERNATIONAL BUREAU To: **PCT** Commissioner **NOTIFICATION OF ELECTION US Department of Commerce** United States Patent and Trademark (PCT Rule 61.2) Office, PCT 2011 South Clark Place Room CP2/5C24 Arlington, VA 22202 Date of mailing (day/month/year) **ETATS-UNIS D'AMERIQUE** 07 February 2001 (07.02.01) in its capacity as elected Office International application No. Applicant's or agent's file reference PCT/GB00/02450 CDK1694 International filing date (day/month/year) Priority date (day/month/year) 22 June 2000 (22.06.00) 24 June 1999 (24.06.99) Applicant HAWKINS, John 1. The designated Office is hereby notified of its election made: in the demand filed with the International Preliminary Examining Authority on: 20 December 2000 (20.12.00) in a notice effecting later election filed with the International Bureau on: 2. The election was not made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland

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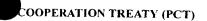
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INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference	FOR FURTHER see Notification (of Transmittal of International Search Report
CDK1694	ACTION (Form PCT/ISA/2	220) as well as, where applicable, item 5 below.
International application No.	International filing date (day/month/year)	(Earliest) Priority Date (day/month/year)
PCT/GB 00/02450	22/06/2000	24/06/1999
Applicant		I
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This International Search Report has been according to Article 18. A copy is being tra	n prepared by this International Searching Authorsmitted to the International Bureau.	nority and is transmitted to the applicant
This International Search Report consists of	of a total of2 sheets.	
	a copy of each prior art document cited in this	report.
Basis of the report		
 With regard to the language, the in 	nternational search was carried out on the bases otherwise indicated under this item.	sis of the international application in the
Authority (hule 23.1(b)).	as carried out on the basis of a translation of th	
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	d unsearchable (See Box I).	
3. Unity of invention is lacki	ng (see Box II).	
4. With regard to the title,		
X the text is approved as sub-	mitted by the applicant.	
	ed by this Authority to read as follows:	
5. With regard to the abstract,		
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the text has been established within one month from the disconnection.	ed, according to Rule 38.2(b), by this Authority date of mailing of this international search repo	as it appears in Box III. The applicant may, ort, submit comments to this Authority.
6. The figure of the drawings to be publish	hed with the abstract is Figure No.	
as suggested by the applica		None of the figures.
because the applicant failed	-	
because this figure better ch	naracterizes the invention.	



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(54) Title: SURFACTANT EMULSIONS AND STRUCTURED SURFACTANT SYSTEMS

(57) Abstract: A mixture of an ethoxylated non-ionic surfactant having an average of 20 to 100 ethylene-oxy groups per molecule with from 10 to 150 parts by weight of a water soluble thiocyanate stabilises non-ionic emulsions and in particular deflocculated non-ionic structured surfactant systems by raising the lamellar/ L_2 phase transition temperature.

SURFACTANT EMULSIONS AND STRUCTURED SURFACTANT SYSTEMS

The present invention relates to non-ionic surfactant emulsions and to the formulation of structured surfactant suspending systems. It is particularly relevant to the formulation of laundry detergents especially those used for industrial and institutional cleaning.

STRUCTURED SURFACTANT

Suspending solids in liquids presents a problem. If the solids differ in density from the liquid they will tend either to sediment or float. Increasing the viscosity of the liquid can retard, but not prevent such separation, and high viscosities are generally undesirable. Colloidal systems, in which the suspended particles are sufficiently small to experience Brownian motion, e.g. less than 1 micron, may be kinetically stable. However the difficulty or undesirability of comminuting some solids to such sizes, and the impossibility of maintaining many of them at this level in the face of crystal growth or agglomeration, limits the use of colloidal suspensions.

Adjusting the density of one phase to match that of the other is usually impracticable. Moreover such systems are almost always temperature-unstable due to differential rates of thermal expansion.

One method of suspension which permits even relatively large particles to be stably suspended is structured surfactant. The term covers systems in which a surfactant mesophase, usually a lamellar or G-phase, alone or more usually interspersed with an aqueous phase, provides a yield stress which is sufficient, when the system is at rest, to immobilise any suspended particles, but which is sufficiently low to allow the system to be poured like a normal liquid. Such systems may display very low apparent viscosities when stirred, pumped or poured and yet be capable of maintaining particles, sometimes of millimetre or larger size, indefinitely in suspension.

Three main types of suspending system have been employed in practice, all involving a G-phase, in which bilayers of surfactant are arranged with the hydrophobic part of the molecule on the interior and the hydrophilic part on the exterior of the bilayer (or vice versa). The bilayers lie side by side, e.g. in a parallel or concentric configuration, Sometimes separated by aqueous layers. G-phases (also known as L_{α} phases) can usually be identified by their characteristic textures under the polarising microscope and/or by x-ray diffraction, which is often able to detect evidence of lamellar symmetry. Such evidence may comprise first, second and sometimes third order peaks with d-spacing ($\frac{2\Pi}{Q}$ where Q is the momentum transfer vector) in a simple integral ratio 1:2:3. Other types of symmetry give different ratios, usually non integral.

Most surfactants form a G-phase either at ambient or at some higher temperature when mixed with water in certain specific proportions. However such G-phases cannot usually be used as structured suspending systems. Useful quantities of solid render them unpourable and smaller amounts tend to sediment.

The main types of structured system used in practice are based on dispersed lamellar, spherulitic and expanded lamellar phases. Dispersed lamellar phases are two phase systems in which the surfactant bilayers are arranged as parallel plates to form domains of G-phases which are interspersed with an aqueous phase to form an opaque gel-like system. They are described in EP O 086 614.

Spherulitic phases comprise well defined spheroidal bodies, usually referred to in the art as spherulites, in which surfactant bilayers are arranged as concentric shells. The spherulites usually have a diameter in the range 0.1 to 15 microns and are dispersed in an aqueous phase in the manner of a classical emulsion, but interacting to form a structured system. Spherulitic systems are described in more detail in EP O 151 884.

Many structured surfactant systems are intermediate between dispersed lamellar and spherulitic, involving both types of structure. Usually systems having a more spherulitic character are preferred because they tend to have lower viscosity. A variant on the spherulitic system comprises prolate or rod shaped bodies sometimes referred to as batonets.

A third type of structured surfactant system comprises an expanded G-phase. It differs from the other two types of structured system in being essentially a single phase, and from conventional G-phase in having a wider d-spacing. Conventional G-phases have a d-spacing of about 5 to 7 nanometers. Attempts to suspend solids in such phases result in stiff pastes which are either non-pourable, unstable or both. Expanded G-phases with d-spacing between 8 and 20, e.g. 10 to 15 nanometers, form when the electrolyte is added to aqueous surfactants at concentrations just below those required to form a normal G-phase, particularly to surfactants in the M phase. The M phase comprises surfactant molecules arranged to form cylindrical rods of indefinite length. It exhibits hexagonal symmetry and a distinctive texture under the polarising microscope. Typical M phases have so high a viscosity that they appear to be curdy solids. M phases near the lower concentration limit (the L₁/M phase boundary) may be pourable but have a very high viscosity and often a mucous-like appearance. Such systems tend to form expanded G-phases particularly readily on addition of sufficient electrolyte.

Expanded G-phases are described in more detail in EP O 530 708. In the absence of suspended matter they are translucent, unlike dispersed lamellar or spherulitic phases which are necessarily opaque. They are optically anisotropic and have shear dependent viscosity. In this they differ from L₁ phases which are micellar solutions and which include microemulsions. L₁ phases are clear, optically isotropic and substantially Newtonian. They are unstructured and cannot suspend solids. Some L₁ phases exhibit small angle x-ray diffraction spectra which show evidence of hexagonal symmetry. Such phases usually have concentrations near the L₁/M phase boundary and may form expanded G-phases on addition of electrolyte.

Most structured surfactant systems require the presence of electrolyte as well as surfactant and water in order to form structured systems capable of suspending solids. However certain relatively hydrophobic surfactants such as isopropylamine alkyl benzene sulphonate can form spherulites in water in the absence of electrolyte. Such surfactants are capable of suspending solids in the absence of electrolyte as described in EP O 414 549.

FLOCCULATION

A problem with the two phase structured surfactant systems, and especially spherulitic systems, is flocculation of the dispersed surfactant structures. This tends to occur at high surfactant and/or high electrolyte concentration. It can have the effect of making the composition very viscous and/or unstable with the dispersed surfactant separating from the aqueous phase.

Certain amphiphilic polymers have been found to act as deflocculants of structured surfactants. One type of deflocculant polymer exhibits cteniform (comb-shaped) architecture with a hydrophilic backbone and hydrophobic side chains or vice versa. A typical example is a random copolymer of acrylic acid and a fatty alkyl acrylate. Cteniform deflocculants have been described in a large number of patents, for example WO-A-9106622.

A more effective type of deflocculant has surfactant rather than cteniform architecture, with a hydrophilic polymer group attached at one end to a hydrophobic group. Such deflocculants are typically telomers formed by telomerising a hydrophilic monomer with a hydrophobic telogen. Examples of surfactant deflocculants include alkyl thiol polyacrylates and alkyl polyglycosides. Surfactant deflocculants are described in more detail in EP O 623 670.

A disadvantage of both surfactant and cteniform deflocculants is that the concentration required to deflocculate to optimum viscosity is critical within fairly narrow limits and varies with temperature. Either too little or too much deflocculant

causes instability and/or excessive viscosity. As a result the deflocculated systems tend to separate if the temperature varies significantly. In particular there is a tendency to form a clear bottom layer on prolonged standing.

One approach to the problem of temperature stability has been to add highly cross linked polyacrylates (see US 5 602 092). These are difficult to disperse in the structure liquid. Our copending application of even date describes the use of certain copolymers to prevent the bottom separation.

APPLICATIONS OF STRUCTURED SURFACTANT

Structured surfactants have been applied to the problems of suspending: water insoluble or sparingly soluble builders in laundry detergent; antifoams and enzymes in laundry detergents and other surfactant systems; abrasives in hard surface cleaners; pesticides and oils in agrochemical preparations (EP O 388 239 and EP O 498 231); rock cuttings in drilling muds (EP O 430 602); dyestuffs in dyebath concentrates and printing inks (EP O 472 089); talcs, oils and other cosmetic ingredients in personal care formulations.

THE PROBLEM

A further kind of temperature instability is often observed with more concentrated structured surfactants especially deflocculated structured surfactants. It typically involves sedimentation of the suspended solid when the composition is stored under warm conditions. We now believe that this separation may be due to a tendency for a phase change in the surfactant from dispersed lamellar or spherulitic to L_2 at elevated temperature.

A similar problem sometimes occurs when attempts are made to prepare non-ionic surfactant emulsions at high concentrations.

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A particular problem arises in relation to liquid detergents suitable for use in industrial and institutional laundries, such as factories, hospitals and hotels and especially in automatically dosed washing machines.

An ideal laundry detergent for institutional use would combine: high surfactant levels and in particular high levels of non-ionic surfactant, which has been found particularly effective for removing soil; high alkalinity, to saponify fatty soil; and high levels of builder, which improve the performance of the surfactant by counteracting the effects of calcium in the water. The composition must be homogeneous and pourable and the concentration as high as possible. Unfortunately it is generally difficult to combine surfactants with electrolytes at high concentrations to form stable compositions. It has proved particularly difficult to achieve this with non-ionic surfactants which are not capable of forming stable solutions at high alkalinity or in the presence of electrolyte, except at very low concentrations which are too low to be commercially acceptable. As a consequence it has hitherto been customary to use two separate solutions in institutional machines, one to supply the surfactant and a separate solution as the source of the alkali.

Attempts to combine the two in a single formulation have hitherto been unsuccessful. Even the use of a defloculant such as those described in EP-A-0 623 670 or EP-A-0 346 995 has not been successful in forming a sufficiently stable homogeneous phase of commercially acceptable concentration, or has done so only over a very restricted temperature range.

THE SOLUTION

We have now discovered that a mixture of a highly ethoxylated non-ionic surfactant and an alkali metal thiocyanate can form highly concentrated emulsions which are temperature stable and also improves the temperature stability of deflocculated structured surfactants. It is readily dispersed in aqueous systems, which are rendered less sensitive to temperature variations thereby.

The mixture is particularly useful for preventing instability in concentrated industrial and institutional detergent. We believe that the mixture inhibits the transition from a G-phase or structured system to an L_2 phase by elevating the phase transition temperature.

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THE INVENTION

Our invention, according to a first embodiment provides a mixture of an ethoxylated non-ionic surfactant having an average of from 20 to 100 ethylene-oxy groups per molecule with from 0.1 to 150 parts by weight of a water soluble thiocyanate.

According to a second embodiment the invention provides a concentrated non-ionic surfactant emulsion comprising water, and said mixture in a concentration adapted to form an emulsion or G phase at a temperature below 40°C

According to a third embodiment our invention provides a structured surfactant composition capable of suspending solids which comprises a structuring surfactant, water and, if required, electrolyte in relative proportions adapted to form a dispersed lamellar and/or spherulitic structured surfactant system, capable of forming an L₂ phase at some temperature below 50°C and, optionally sufficient of a deflocculant to inhibit the flocculation of said system <u>characterised in that</u> said composition comprises an effective amount of a phase stabiliser which is a mixture of (i) an ethoxylated non-ionic surfactant having from 20 to 100 ethylene oxy groups and (ii) a water soluble thiocyanate in a relative molar proportion (i):(ii) of from 1:200 to 20:1.

Preferably said structuring surfactant consists of a major amount of non-ionic surfactant, typically an ethoxylate with 1 to 15 e.g. 2 to 10 ethylene oxide and optionally a minor amount of anionic and/or amphoteric surfactant. Preferably said water is present in a proportion of from 20 to 60%. Preferably said electrolyte comprises alkali.

THE PHASE STABILISER

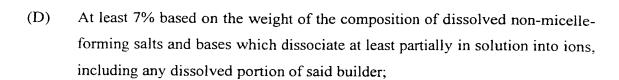
The stabiliser may comprise an ethoxylated $C_{8\cdot20}$, straight or branched chain alcohol or fatty acid, fatty amine, sorbitan or glycerol ester, alkyl polypropoxy group or alkyl phenyl group. The number of ethoxy groups may be from 20 to 100, e.g. 30 to 80, preferably 40 to 60. The mole ratio of (i):(ii) may preferably be from 1:100 to 10:1, e.g. 1:50 to 5:1. When added to a structured surfactant system we prefer that the surfactant (i) be present in an amount of from 0.1 to 3% by weight of the total composition, preferably 0.2 to 2%. The maximum concentration depends on the amount of the structuring surfactant present, higher levels being preferred for higher concentrations of structuring surfactant. Excessive amounts break structures. The thiocyanate is preferably present in an amount greater then 0.05%, more preferably greater than 0.1%, e.g. greater than 0.5%. The upper limit is not critical but concentrations greater than about 10% are unlikely to provide additional benefit. Concentrations greater than 2% are generally uneconomic.

The thiocyanate may be any water soluble thiocyanate but is preferably an alkali metal or ammonium thiocyanate and most preferably potassium thiocyanate.

INSTITUTIONAL AND INDUSTRIAL FORMULATIONS

According to a preferred embodiment the invention provides a detergent composition comprising:-

- (A) 10 to 50% by weight of the composition of water;
- (B) At least 3% based on the weight of the composition, preferably 4 to 10%, of a structured surfactant comprising more than 50% based on the total weight of surfactant of non-ionic surfactants having a mean HLB of from 8 to 15 and optionally a smaller proportion of anionic and/or amphoteric surfactant;
- (C) At least 10% by weight based on the weight of the composition of builders;



- (E) A total free alkalinity of at least 0.5 normal;
- (F) Sufficient of a deflocculant to provide, in conjunction with components A to E above a pourable composition which does not separate after 1 month at 25°C;

Wherein there is additionally from 0.01% to 5% by weight of an auxiliary stabiliser consisting of C_{8-20} alcohol ethoxylates having an average of from 25 to 100 ethyleneoxy groups per molecule, together with a water soluble, preferably potassium, thiocyanate.

The amount of water is typically greater than 15%, preferably greater than 20%, especially greater than 25% and usually greater than 30% based on the total weight of the composition.

The structuring surfactant is preferably all non-ionic since in some applications inclusion of anionic surfactant can adversely affect performance. However where anionic surfactant can be tolerated its inclusion has the advantage of enabling higher total concentrations of surfactant to be more easily achieved. Typically wholly non-ionic based formulations contain from 7 to 30%, more typically 10 to 25% by weight surfactant while compositions containing a minor proportion of anionic surfactant may contain up to 50% by weight, e.g. 15 to 40%, especially 20 to 35%.

The non-ionic surfactant preferably consists of from 60 to 100% by weight of alkoxylate, preferably ethoxylate or mixed ethoxylate/propoxylate. Typically it comprises alkoxylated C_{8-20} , especially C_{10-18} natural or synthetic alcohols. The

alcohols are typically primary or secondary, straight or branched chain, saturated or unsaturated. Also effective are alkoxylated fatty acids, fatty amines, alkyl phenols, glyceryl mono and dialkyl esters and sorbitan esters.

The ethoxylate typically contains an average of from 1 to 10 alkoxy groups depending on the alkyl chain length, to give an HLB of from 10 to 15, preferably 12 to 14.

The non-ionic surfactant may comprise a mono- or di- ethanolamide or an amine oxide. The surfactant may optionally contain a minor proportion (i.e. less than 50% based on the total weight of surfactant) of anionic surfactant such as soap and/or alkyl benzene sulphonate. Other anionic surfactants which may be used include alkyl ether sulphates, alkyl sulphates, olefin sulphonates, paraffin sulphonates and alkyl phosphates.

The builder is preferably sodium tripolyphosphate, but may alternatively be or comprise sodium or potassium pyrophosphate, sodium or potassium citrate, sodium or potassium carbonate or a zeolite. Other builders include EDTA, nitrilotriacetate, phosphonates and poly electrolytes such as polyacrylates or polymaleates. The term "builder", as used herein, excludes any hydroxides used to provide the free alkali but includes carbonate and silicate. The builder is present in amounts greater than 10% by weight based on the total weight of the composition, preferably more than 15%. Levels of builder may be above 20%, any excess over the solubility in the system being present as suspended particles. Builder concentrations do not normally exceed 50% by weight and are usually less than 40%, e.g. less than 30%.

The composition contains a total of at least 7% by weight of dissolved surfactant desolubilising salts and bases. This includes any dissolved portion of the builder and any alkali required to provide the free alkalinity.

It excludes micelle forming components such as anionic surfactant. The dissolved salts and bases preferably constitute from 10 to 40%, e.g. 15 to 30% by weight of the composition. and sufficient to form a multiphase system in which an aqueous phase is interspersible with a surfactant or surfactant mesophase.

The total free alkali should be sufficient to neutralise at least an equal volume of 0.5 normal HCl. Preferably the free alkalinity is from 0.7 to 2 normal, e.g. 0.8 to 1.5.

We particularly prefer that compositions of the invention contain a deflocculant. The deflocculant may be a polycarboxylate having on or more alkyl groups such as C_{8-20} alkyl thiol polycarboxylate e.g. polyacrylate or polymaleate, or a copolymer of unsaturated carboxylic acid with a C_{8-20} alkyl ester of an unsaturated carboxylic acid e.g. a copolymer of acrylic acid and/or maleic acid with a minor proportion of a C_{8-20} alkyl acrylate and/or alkyl maleate ester. Alternatively it may comprise an alkyl polyglycoside. The alkyl polyglycoside is preferably a polyglucoside and typically has an average degree of polymerisation between 1.3 and 10, more usually 1.5 to 3.

The deflocculant is generally added in an amount sufficient to provide an interspersion of the aqueous phase with the surfactant phase at 25°C, which does not separate within 1 month. This may typically require from 0.5 to 10, more usually 1 to 5%, e.g. 2 to 4.5% by weight based on the weight of the composition. The amount is preferably adjusted to obtain a spherulitic composition comprising surfactant vesicles, usually having a multilamellar or G-phase structure, dispersed in an aqueous phase.

The auxiliary stabiliser may be present in proportions up to 5% by weight, usually 0.01 to 3%, e.g. 0.02 to 2 especially 0.01 to 1. Combinations of two or more auxiliary stabilisers may sometimes be particularly effective.

The detergent compositions of the invention preferably also contain the conventional minor detergent ingredients including antifoams such as silicone antifoam, soil suspending agents such as a carboxymethyl cellulose, optical brighteners, stain removers such as enzymes, bleaches including perborate metaborate mixtures,

sequestrants such as phosphonates and especially amino phosphonates including aminotrismethylene phosphonate, ethylene diamine tetrakis (methylene phosphonate), diethylene triamine pentakis (methylene phosphonate) and others in the same series, perfumes, colouring, preservatives, corrosion inhibitors, bleach activators such as TAED and/or fabric conditioner.

The aforesaid minor ingredients may all be present in conventional amounts and usually constitute a total of less than 5% by weight of the composition, typically less than 1%. The anionic component of the ionic ingredients may typically be sodium, potassium or a mixture of the two. Potassium is preferred where very high solids contents are desired.

The invention is illustrated by the following examples in which all proportions are by weight of the 100% material based on the weight of the composition.

	% wt/wt
Potassium thiocyanate	0.5
Optical brightener "TINOPAL" ® CBS/X	0.1
C ₁₂₋₁₄ alkyl (polyglucoside (dip.=1.4)	2.25
Sodium tripolyphosphate	19.0
C ₁₂₋₁₈ alkyl fifty mole ethoxylate	0.4
C ₁₂₋₁₄ branched alkyl seven mole ethoxylate	11.0
Sodium hydroxide	10.0
Water	balance

The above product was an effective industrial and institutional laundry detergent. In the absence of the alkyl polyglycoside the composition was heavily flocculated and underwent rapid separation. In the absence of the potassium thiocyanate and/or the fifty mole ethoxylate the product separated at temperatures above 30°C.

CLAIMS

- 1. A mixture of an ethoxylated non-ionic surfactant having an average of from 20 to 100 ethylene-oxy groups per molecule with from 0.1 to 150 parts by weight of a water soluble thiocyanate.
- 2. A concentrated non-ionic surfactant emulsion comprising water, and said mixture in a concentration adapted to form an emulsion or G phase at a temperature below 40°C.
- 3. A structured surfactant composition capable of suspending solids which comprises a structuring surfactant, water and, if required, electrolyte in relative proportions adapted to form a dispersed lamellar and/or spherulitic structured surfactant system, capable of forming an L₂ phase at some temperature below 50°C and, optionally sufficient of a deflocculant to inhibit the flocculation of said system characterised in that said composition comprises an effective amount of an phase stabiliser which is a mixture of (i) an ethoxylated non-ionic surfactant having from 20 to 100 ethylene oxy groups and (ii) a water soluble thiocyanate in a relative molar proportion (i):(ii) of from 1:200 to 20:1.
- 4. A composition according to claim 3 wherein said structuring surfactant comprises a major amount of a non-ionic surfactant and a minor amount of anionic and/or amphoteric surfactant.
- 5. A composition according to either of claims 1 and 2 wherein said stabiliser comprises an ethoxylated C₈₋₂₀ fatty alcohol.

- 6. A detergent composition comprising:-
- (A) 10 to 50% by weight of the composition of water;
- (B) At least 3% based on the weight of the composition, of a structured surfactant comprising more than 50% based on the total weight of surfactant of non-ionic surfactants having a mean HLB of from 8 to 15 and optionally a smaller proportion of anionic and/or amphoteric surfactant;
- (C) At least 10% by weight based on the weight of the composition of builders;
- (D) At least 7% based on the weight of the composition of dissolved non-micelleforming salts and bases which dissociate at least partially in solution into ions, including any dissolved portion of said builder;
- (E) A total free alkalinity of at least 0.5 normal;
- (F) Sufficient of a deflocculant to provide, in conjunction with components A to E above a pourable composition which does not separate after 1 month at 25⁰C;

Wherein there is additionally from 0.01% to 5% by weight of an auxiliary stabiliser consisting of C_{8-20} alcohol ethoxylates having an average of from 25 to 100 ethyleneoxy groups per molecule, together with a water soluble thiocyanate.

7. A composition according to claim 6 wherein thiocyanate is potassium thiocyanate.

INTERNATIONAL SEARCH REPORT



A CLASS	FICATION OF SUBJECT MATTER		GB U	0/02450
IPC 7	C11D17/00 C11D1/72 C11D3	/04		
1	to International Patent Classification (IPC) or to both national class	sification and IPC		
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9	ta, EPO-Internal, PAJ, CHEM ABS Da		search terms use	a)
C. DOCUM	ENTS CONSIDERED TO BE RELEVANT			
Category °	Citation of document, with indication, where appropriate, of the	relevant passages		Relevant to claim No.
Α	EP 0 079 646 A (UNILEVER) 25 May 1983 (1983-05-25) abstract; example 3			2-5
A	EP 0 414 549 A (ALBRIGHT & WILS 27 February 1991 (1991-02-27) cited in the application example 1	ON)		2-5
A	DATABASE WPI Section Ch, Week 199548 Derwent Publications Ltd., Lond Class G04, AN 1995-366904 XP002149038 & CN 1 096 010 A (LIAO S), 7 December 1994 (1994-12-07) abstract	on, GB;		1
Furthe	er documents are listed in the continuation of box C.	X Patent family me	mbers are listed in	n annex.
° Special cate	egories of cited documents :	"T" later document publish	and after the inter-	national filing data
"A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier document but published on or after the international "T" later document published after the inter or priority date and not in conflict with a circumstant of the understand the principle or the inventor.			he application but ory underlying the	
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